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(71) Applicant *(for all designated States except US)*: **NOTE PRINTING AUSTRALIA LIMITED** [AU/AU]; Hume Highway, Craigieburn, VIC 3064 (AU).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **GHIOGHIU, Ana** [AU/AU]; 38 Hopetoun Street, Elsternwick, VIC 3185 (AU). **HARDWICK, Bruce, Alfred** [AU/AU]; 5 Scanlons Road, Wandong, VIC 3758 (AU). **BATISTATOS, Odi** [AU/AU]; 28 Volga Street, Glenroy, VIC 3046 (AU). **NEMETH, Joshua** [AU/AU]; 8 Portsmouth Street, Heathmont, VIC 3135 (AU).

(74) Agent: **FREEHILLS CARTER SMITH BEADLE**; 101 Collins Street, Melbourne, VIC 3000 (AU).

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(54) Title: INTAGLIO PRINTING INKS

(57) Abstract: An Intaglio printing ink is disclosed that uses as a binder an autoxidisable polyester resin. The fatty acid content is in the range 47 - 52% w/w and the preferred fatty acid is linseed fatty acid. The acid value of the resin in the range 50 - 70 mg KOH/g. The printing ink includes solvents and pigments and can be cured under UV irradiation to produce useful printed material on polymer substrates.

INTAGLIO PRINTING INKS

Technical Field

This invention relates to printing inks and in particular to printing inks known as Intaglio printing inks. The invention also relates to a process of obtaining a dry Intaglio printing ink film on a substrate.

Background of the Invention

Intaglio printing is a process whereby an image is transferred to a sheet of paper or other substrate from a plate with recessed portions, where printing ink is located in the recessed portions of the plate. After transfer of the printing ink to the paper or other substrate the ink may be dried either under ambient conditions or under forced drying conditions of elevated temperatures and/or increased air flow. The printing ink also dries or cures by undergoing chemical reactions.

The Intaglio technique is well known and produces high quality raised images that can be used for the manufacture of banknotes and other documents as well as for artworks. One of the features of the Intaglio printing technique is that the film build of the printing ink that is transferred to the paper or other substrate may vary from a few microns to fifty microns or more. This ability to vary the film build is a desirable feature of Intaglio printing and may be used to advantage to produce embossing and other desirable features such as tactility without embossing. This feature also distinguishes inks suitable for use as Intaglio inks from other printing inks.

The high film build characteristic of Intaglio printing accentuates the problem of "set-off". "Set-off" is the transfer of ink from one finished sheet to the next finished sheet in the run. It is caused by residual tackiness on the surface of the printed sheet and often is exacerbated when pressure is applied by the weight of a pile of stacked sheets. "Set-off" is usually worst on sheets towards the bottom of the stack as these are under the most weight . One method used to reduce the problem of "set-off" is to interleave sheets between adjoining sheets. However interleaving is a very cumbersome and costly process and not desirable for commercial printing operations. The type of substrate can also affect "set-off". The "set-off" problem tends to be worse with substrates that cannot readily absorb

solvents from the printing ink. In the case of banknotes, the "set-off" is generally worse when polymer substrates are used compared to cellulosic natural product paper substrates.

EP 486092 (Akzo) discloses fatty acid resinous derivatives of starch type polyether polyols. The acid value of the resins have acid values of about 10 mgKOH/g. There is no suggested use as Intaglio inks.

EP 750026 (Sun Chemical) relates to Intaglio printing inks. The resins disclosed in that citation are addition polymerised modified epoxy resin fatty acid esters. The epoxy resin is bisphenol A type resin which are polyethers. After application, the inks are thermally cured.

It is an object of the present invention to improve the "set-off" of Intaglio printing inks, especially on polymer substrates.

Summary of the Invention

This invention provides in one form an Intaglio ink wherein the binder includes an autoxidisable polyester resin with a fatty acid content in the range 40 – 60% by weight, more preferably 45 – 55% and most preferably 47 – 52%.

Preferably the fatty acid has at least 25% of its chain as linolenic acid, more preferably at least 40%. Preferably the iodine value of the fatty acid is at least 150gl/100g. Preferably the fatty acid is selected from the group consisting of linseed fatty acid, tung oil fatty acid or synthetic equivalents. Preferably the fatty acid is linseed fatty acid.

Preferably the acid value of the resin is in the range 40 – 80 mg KOH/g of resin and more preferably 50 – 70 mg KOH/g of resin and most preferably 55 – 65 mg KOH/g of resin.

Preferably the resin has a viscosity as measured by Gardner-Holt viscosity bubble tubes in the range R – Y and more preferably T – X and most preferably T – W when measured as a 60% solution in ethyl di-icinol (monoethyl ether of diethyleneglycol).

This invention provides in an alternative form an Intaglio ink which comprises:

20 – 40% an autoxidisable polyester resin

10 – 30% organic solvents

1 – 60%, preferably 1 – 50% pigments

0 – 5% catalysts

wherein the autoxidisable polyester resin is as described above.

Preferably the ink composition further comprises a wax dispersion.

The glass transition temperature (Tg) of the wax dispersion is preferably less than the maximum temperature achieved during the printing process and more preferably more than 20°C less than the maximum temperature achieved during the printing process. Suitable wax dispersions include polyurethane dispersions and micronised polyolefin dispersions.

The invention provides in a further form a method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as described above, exposing the film to a UV light source and curing the ink.

The solvent present in the Intaglio printing ink composition may consist of a single solvent. However it is preferred that a mixture of solvents is used. Different solvents may be used to influence various properties of the ink composition, including viscosity, ink stability and transfer properties. Solvents that are released reasonably quickly from the inked substrate may be useful as their release will tend to aid the time to achieve satisfactory "set-off". Illustrative of such solvents are aliphatic type blends of hydrocarbons that have boiling points in the range 150–270°C. Slower solvents may also be used in conjunction with these hydrocarbon solvents and these other solvents may be retained to a certain extent after the ink has dried. Effectively these solvents, at their residual levels, can function as plasticisers and provide greater levels of flexibility to the dried ink. Examples of such solvents are glycols and ether - glycols. Good flexibility in the dried ink is particularly preferred with bank notes where the notes are frequently subjected to bending, flexing and folding.

Driers that may be used in the ink compositions of the present invention include the transition metal salts commonly used in accelerating the drying of alkyd or oil based house paints. A particularly preferred drier is a combination of cobalt

and manganese. These metals are usually in the form of soluble or compatible salts such as, for example, cobalt naphthenate or cobalt octoate. Some of the metal driers facilitate surface cure while others enhance through cure. In certain embodiments of the present invention it is believed to be desirable to achieve a greater amount of surface cure compared to through cure. It is believed this surface cure will lead to the desired "set-off" property while the lack of extensive through cure will assist in increasing the flexibility of the film.

The pigments that may be used in the printing ink compositions of the present invention include those commonly used in printing inks. Examples include organic pigments, carbon black and iron oxide. The levels of pigment present may vary depending on the particular application but are generally in the range of 5 – 60% by weight. Pigments also include fillers and extenders.

In practice the pigments may be dispersed using the Intaglio ink resin as a pigment dispersant. This resin functions effectively as a pigment dispersant and this is believed to be a consequence of the relatively high acid value of the resin. However a separate pigment dispersant may be used and in this case the pigment dispersant would be an additional ingredient in the Intaglio printing ink composition.

Preferably the acid value of the resin is in the range 40 – 80 mg KOH/g of resin and more preferably 50 – 70 mg KOH/g of resin and most preferably 55 – 65 mg KOH/g of resin.

Preferably the resin has a viscosity as measured by Gardner-Holt viscosity bubble tubes in the range R – Y and more preferably T – X and most preferably T – W when measured as a 60% solution in ethyl di-icinol (monoethyl ether of diethyleneglycol). These viscosities indicate relatively high molecular weight and branched chain networks. These are believed to contribute to the desired "set-off" behaviour.

In an alternative form the Intaglio ink further comprises a photoinitiator.

In a still further alternative form this invention provides a process of preparing a dry Intaglio printing ink on a substrate, the process comprising the steps of transferring an Intaglio printing ink from a plate to a substrate at a temperature in the range 60 –

90°C, more preferably 80 – 84°C, irradiating the printed substrate from a UV radiation source.

Detailed Description of the Invention

The use of UV curable coatings has been known for many years. A particularly well known example is that of any clear finishes over timber substrates. These timber substrates are coated in a factory. These precoated timber materials may then be used for flooring or other applications where further coating is not required. UV curable coatings are particularly applicable to clear, unpigmented coatings. The presence of pigments tends to limit the penetration of UV radiation and hence has not been as widely used in pigmented coatings. The general composition of UV coatings includes resins or polymers as binders that have addition polymerisable ethylenic unsaturation. Typically these resins or polymers as well as oligomers or monomers, have multi-functional addition polymerisable unsaturation. Examples include polyols that have been modified as polyacrylates, such as trimethylolpropane triacrylate, and pentaerythritol tri and tetra acrylate. Photoinitiators are commonly used in such compositions to accelerate the rate of cure or crosslinking.

Polyester resins or polymers that include fatty acid side chains are well known in coating technology. They are referred to as alkyds. They are used as the binders in household "oil" paints. Coatings prepared using alkyd resins as binders form films both by evaporation of solvent as well as chemical reaction utilising oxygen in the atmosphere. They are generally referred to as autoxidisible coatings as a result of the mechanism of curing involving atmospheric oxygen. Generally the fatty acids that are used as binders in household coatings are derived from naturally occurring soya or safflower oils. Linseed fatty acids are generally not used because of problems with yellowing.

The percentage of fatty acid present is usually about 60% by weight. Typical alkyd based coatings take 2- 4 hours to achieve tack free condition. While lower levels of fatty acid lead to improve "tack-free" times the best balance of properties is achieved at about the 60% level of fatty acid. Lower levels of fatty acids generally require the presence of aromatic solvents which are undesirable for

domestic applications. Generally improved "tack-free" times are achieved with lower percentages of fatty acids and these binders are used in industrial applications. "Tack-free" is generally achieved in such applications by evaporation of solvent alone. The mechanism of curing alkyd based coatings has been widely studied. While the fatty acid side chains have unsaturated groups it is generally recognised that the mechanism of cure does not involve addition polymerisation reactions with this unsaturation. In contrast the binders for UV coatings generally require the presence of addition polymerisable ethylenic unsaturation. Accordingly, it would not be expected that alkyd based coatings would be susceptible to curing reactions in response to UV radiation. Furthermore, the autoxidation type mechanisms would be anticipated to be too slow to lead to acceptable "set-off" times when used as a printing ink.

The polyester resin of the present invention may be prepared by known methods that are applicable to the preparation of alkyds. The composition of the polyester binders of the present invention may be varied within the parameters outlined in the Summary of the Invention. It is preferred that the acid value of the resins is relatively high and it is believed this contributes to good adhesion to the substrate, particularly when these are polymer substrates used for bank notes. As discussed previously, it also aids pigment dispersion The polyester resin of the present invention preferably includes at least 20% by weight of aromatic polyfunctional carboxylic acids and it is believed this contributes to a relatively high Tg of the polyester backbone. This relatively high aromatic content is believed to assist in obtaining satisfactory "set-off". While the resins are described as polyesters other minor levels of non ester groups may be incorporated. However the polymer backbone of the resin is required to be essentially a polyester. Minor amounts of amide or ether groups could also be included.

The printing inks of the present invention may be used conventionally, that is without UV radiation. However, generally improved results are achieved when they are subjected to UV radiation. Typically when used conventionally "set-off" is adequate to allow 250 sheets to be stacked. However, with UV irradiation 2,000 – 3,000 sheets may be stacked. Generally, the Intaglio printing is carried out at 80 –

84°C as a primary source of heat, and curing/drying continues afterwards at normal temperatures. The curing may be accelerated with other radiation. We have found that by using a combination of heated plates and a UV system incorporating exhaust ventilation, acceptable initial cure of the ink is achieved. Generally UV systems have a significant infra-red component which assists thermal curing/drying. In this alternative embodiment involving UV curing photoinitiators may also be used. These initiators that are well known in the UV curing of coatings, for example Irgacure 184 (ex Ciba-Geigy).

As well as the components outlined in the Summary of the Invention, other materials may be included without detracting from the overall properties of the printing ink. As an example, fillers or extenders can be used to vary the gloss of the printing ink image. These are included within the pigment category and include calcium carbonate as an example. The driers or catalysts that may be used are well known. Some driers are known more as "surface driers", while other driers are known as "through driers". The selection of driers for the present invention may be made to obtain the requisite balance of properties. It is believed desirable in many banknote applications that "through dry" is limited. This ensures adequate flexibility of the printing ink. It is believed that the UV irradiation accentuates the "surface cure" and hence to an extent limits the "through cure". Minor amounts of other binders as well as plasticisers may be used without detracting from the properties of the compositions of the present invention. While the printing inks of the present invention are particularly suitable for plastic substrates, they can also be used on other substrates such as cellulosic type substrates. The compositions are particularly useful on polymer substrates used for security documents such as banknotes.

The types of apparatus that may be used to irradiate the sheets of substrate may vary but generally involve the exposure of a substrate to which the printing ink has been applied for a short period of time with UV radiation.

The invention will be further described by reference to a preferred embodiment in the following examples.

Example 1

Preparation of an Intaglio resin according to the invention.

The following reagents were charged to a three litre flanged top flask equipped with mechanical stirrer, condenser, nitrogen sparge and electric mantle.

ethylene glycol	168g	
fatty acid	882g	A
trimethylol propane	262g	B
terephthalic acid	202g	C
isophthalic acid	202g	D
dibutyl tin oxide	0.7g	E
styrene maleic anhydride resin	104g	F
trimellitic anhydride	179g	G
isopar G	636g	H

Stage A was charged to the stirred reactor and B to E stages were added in that order.

The reactants were heated to 182°C and maintained at that temperature until 66ml of water was removed. The refractive index of the collected water was monitored to ensure no loss of ethylene glycol. The column temperature was not allowed to exceed 110°C.

The batch was then heated to 260°C again ensuring that the column head temperature was maintained below 110°C. After a total of 104 mls of water had been collected the acid value was monitored until it was in the range 6 – 9 mg KOH/g and the batch was clear. The Gardner Holt viscosity was in the range A - C as measured in a 60% solution in ethyl di-icinol. Total water removed was 123ml. F stage was then added and the batch maintained at 260 – 265°C until the AV was in the range 6 – 9 and the viscosity was G - I (using the same method as used previously). The batch was cooled to 170°C and G stage was added. The temperature was maintained at 170°C until the acid value was in the range 55 – 65 and the viscosity was in the range T - W. H stage was then added to give the final resin. The fatty acid content of this resin was 44%.

Example 2

This example illustrates the preparation and testing of an Intaglio printing ink composition.

Resin (from Example 1)	10.0
SiO_2 (thickener)	0.6
Irgacure 184 (Ciba-Geigy)	0.2
Co/Mn octoate (6% solution)	0.1
black ink (NNS invis)	0.5

The printing ink composition was prepared by combining the above ingredients. The ink was then tested by exposing a film of thickness 50 μm thickness to 150 Watts/cm of UV irradiation at a distance of 75mm.
The films were then assessed for set-off after passes of the film across the UV radiation source. After five passes the film was rated as very good while after four passes it was rated as fair.

Example 3

Example 2 was repeated but with a resin similar to Example1 except that the fatty acid content was 49% rather than 44%. When assess as in Example 2 it was rated as excellent after either four or five passes. Even after three passes it was rated as very good. After two passes it was rated as fair. Although the fatty acid content of this example was higher than Example 2, the "set-off" was improved. This is contrary to the expected result for "tack-free" behaviour for alkyds.

Claims Defining the Invention are as Follows:

1. An Intaglio ink wherein the binder includes an autoxidisable polyester resin with a fatty acid content in the range 40 – 60% by weight.

2. An Intaglio ink as defined in claim 1 wherein the fatty acid content is in the range 45 – 55%.

3. An Intaglio ink as defined in claim 2 wherein the fatty acid content is in the range 47 – 52%.

4. An Intaglio ink as defined in any one of claims 1 to 3 wherein the acid value of the polyester resins is in the range 40 – 80 mg KOH/g of resin.

5. An Intaglio ink as defined in claim 4 wherein the acid value of the polyester resins is in the range 50 - 70 mg KOH/g of resin.

6. An Intaglio ink as defined in any one of claims 1 to 3 wherein the acid value of the polyester resins is in the range 55 – 65 mg KOH/g of resin.

7. An Intaglio ink as defined in any one of claims 1 - 6 wherein the fatty acid has at least 25% of its chains as linolenic acid.

8. An Intaglio ink as defined in claim 7 wherein the fatty acid has at least 40% of its chains as linolenic acid.

9. An Intaglio ink as defined in any one of claims 1 – 6 wherein the iodine value of fatty acid is at least 150gl/100g.

10. An Intaglio ink as defined in any one of claims 1 – 9 wherein the fatty acid is linseed fatty acid.

11. An Intaglio ink as defined in any one of claims 1 – 10 wherein the resin has a viscosity, as a 60% w/w solution in monoethylether of diethylene glycol, as measured by Gardner-Holt viscosity bubble tubes in the range R – Y.

12. An Intaglio ink as defined in claim 11 wherein the resin has a viscosity as measured by Gardner-Holt viscosity bubble tubes in the range T – X.

13. An Intaglio ink as defined in claim 12 wherein the resin has a viscosity as measured by Gardner-Holt viscosity bubble tubes in the range T – W.

14. An Intaglio ink which comprises:

20 – 40% an autoxidisable polyester resin

10 – 30% organic solvents

1 – 60% pigments

0 – 5% catalysts

wherein the autoxidisable polyester resin is as defined in any one of claims 1 – 13.

15. An Intaglio ink as defined in claim 14 further comprising a wax dispersion.

16. An Intaglio ink as defined in claim 15 wherein the glass transition temperature (Tg) of the wax dispersion is less than the maximum temperature achieved during the printing process.

17. An Intaglio ink as defined in claim 16 wherein the glass transition temperature (Tg) of the wax dispersion is more than 20°C less than the maximum temperature achieved during the printing process.

18. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 1, exposing the film to a UV light source and curing the ink.

19. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 2, exposing the film to a UV light source and curing the ink.

20. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 3, exposing the film to a UV light source and curing the ink.

21. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 4, exposing the film to a UV light source and curing the ink.

22. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 5, exposing the film to a UV light source and curing the ink.

23. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 6, exposing the film to a UV light source and curing the ink.

24. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 7, exposing the film to a UV light source and curing the ink.

25. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 8, exposing the film to a UV light source and curing the ink.

26. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 9, exposing the film to a UV light source and curing the ink.

27. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 10, exposing the film to a UV light source and curing the ink.

28. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 11, exposing the film to a UV light source and curing the ink.

29. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 12, exposing the film to a UV light source and curing the ink.

30. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 13, exposing the film to a UV light source and curing the ink.

31. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 14, exposing the film to a UV light source and curing the ink.

32. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 15, exposing the film to a UV light source and curing the ink.

33. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 16, exposing the film to a UV light source and curing the ink.

34. A method for preparing an Intaglio printed substrate by applying a film of Intaglio ink as defined in claim 17, exposing the film to a UV light source and curing the ink.

35. An Intaglio printed substrate as prepared according to claim 18.

36. An Intaglio printed substrate as prepared according to claim 19.

37. An Intaglio printed substrate as prepared according to claim 20.

38. An Intaglio printed substrate as prepared according to claim 21.

39. An Intaglio printed substrate as prepared according to claim 22.

40. An Intaglio printed substrate as prepared according to claim 23.

41. An Intaglio printed substrate as prepared according to claim 24.

42. An Intaglio printed substrate as prepared according to claim 25.

43. An Intaglio printed substrate as prepared according to claim 26.

44. An Intaglio printed substrate as prepared according to claim 27.

45. An Intaglio printed substrate as prepared according to claim 28.

46. An Intaglio printed substrate as prepared according to claim 29.

47. An Intaglio printed substrate as prepared according to claim 30.

48. An Intaglio printed substrate as prepared according to claim 31.

49. An Intaglio printed substrate as prepared according to claim 32.

50. An Intaglio printed substrate as prepared according to claim 33.

51. An Intaglio printed substrate as prepared according to claim 34.

52. An Intaglio printed substrate as prepared according to claim 35.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU00/01421

A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl. [7]: C09D 11/06, 11/08, 11/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: C09D 11/-

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPAT & JAPIO: ink, polyester, fatty acids

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Derwent Abstract Accession No. 98-280514/25, Class A97, G02, JP 10095945 A (NISSHIN OIL MILLS LTD) 14 April 1998 See Abstract	1-52
X	US 4107012 A (FUHR et al.) 15 August 1978 See column 1 line 56 to column 3 line 17 and Examples 5-6	1-52
X	US 5569701 A (MOYNIHAN) 29 October 1996 See column 4 lines 27-66 and column 5 lines 30-65	1-52

[X] Further documents are listed in the continuation of Box C [X] See patent family annex

* Special categories of cited documents:

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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Date of the actual completion of the international search	Date of mailing of the international search report
6 December 2000	12 DEC 2000
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaustralia.gov.au Facsimile No. (02) 6285 3929	Authorized officer **ALBERT S. J. YONG** Telephone No : (02) 6283 2160

INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU00/01421

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5178672 A (MILLER) 12 January 1993 See column 3 line 46 to column 4 line 15, and Examples	1-52
X	US 3557691 A (BAYER) 26 January 1971 See column 2 lines 48-63 and Examples 1-3	1-52
X	Derwent Abstract Accession No. 92-027493/04, Class A97, G02, JP 03273068 A (TOYO INK MFG KK) 4 December 1991 See Abstract	1-52

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/AU00/01421

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member					
JP	10095945	NONE					
US	4107012	AT	3767/73	BE	798808	CA	1019092
		CH	581675	DE	2221335	ES	414187
		FR	2183053	GB	1398032	IT	986078
		JP	49047483	NL	7305862		
US	5569701	AU	54642/96	CA	2177804	EP	745652
		JP	9100431	NZ	286712		
US	5178672	CA	2043914				
US	3557691	NONE					
JP	03273068	NONE					

END OF ANNEX